

Polarized nuclear target based on parahydrogen induced polarization

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Abstract

We discuss a novel concept of a polarized nuclear target for accelerator fixed-target scattering experiments, which is based on parahydrogen induced polarization (PHIP). One may be able to reach a 33% free-proton polarization in the ethane molecule. The potential advantages of such a target include operation at zero magnetic field, fast (~ 100 Hz) polarization reversal, and operation with large intensity of an electron beam.

I. INTRODUCTION

A. Motivation

Polarized nuclear targets, recently reviewed in Refs. [1,2], play an important role in particle physics in investigations of fundamental symmetries and dynamical aspects of known interactions, in particular, those aimed at studying the physics of the nucleon structure functions³. The structure functions g_1 and g_2 for both proton and neutron are important for understanding of the quantum chromodynamical (QCD) nature of the nucleon spin. Currently, a number of accelerator based scattering experiments explore polarization observables for the structure-function investigations^{3,4}. A new way to study the nucleon spin emerged in the last decade: the SIDIS (semi-inclusive deep inelastic scattering with transversely polarized nucleons) experiments are probing the effect of nucleon polarization on the azimuthal variation of the leading meson-production cross section in deep inelastic electron-nucleon scattering. This effect will allow measurement of the quark orbital angular momentum (OAM), as discussed by Sivers⁵ and Collins⁶. A nonzero value of OAM means that quarks move coherently, at least a fraction of the time, around the spin direction. The concept of OAM is attractive, however, reliable SIDIS data are needed to validate it. While important measurements have been performed at the Deutsches Elektronen Synchrotron (DESY)^{7,8} and the European Organization for Nuclear Research (CERN)^{9,10}, the experimental accuracy needs to be improved by orders of magnitude. The Thomas Jefferson National Accelerator Facility (JLab) at Newport News, Virginia has a large program of SIDIS experiments with a 12 GeV electron beam¹¹. The hadron facility FAIR at Darmstadt, Germany has a physics program for studies of the antiproton interaction with polarized proton and neutron targets¹².

As far as weak interactions are concerned, a parity non-conserving term in deep inelastic electron-scattering (DIS) cross section appears for a polarized electron beam due to the interference of electromagnetic and weak-neutral-current processes ($\gamma - Z$ interference). It was first observed at the Stanford Linear Accelerator Center (SLAC) with an unpolarized target (see Ref.¹³ and references therein). Complementary parity-violating asymmetries arise for an unpolarized beam and a longitudinally polarized target, as predicted in Ref.¹⁴. However, these are hard to measure because the event rates in polarized-target experiments are usually low. This effect potentially provides access to the parity violating structure

functions⁴, which were measured in deep inelastic neutrino/antineutrino-nucleon scattering and high momentum transfer electron-proton DIS¹⁵. It will be interesting to verify these using an unpolarized electron beam and a polarized nucleon target.

B. Polarized nuclear targets

The key parameters of a nuclear target are: the areal density of the target material (N , in nucleons/cm²), the usable beam intensity (I , in electrons/s), the electron-nucleon luminosity, which is the product of these quantities

$$L = N \times I, \quad (1)$$

usually given in Hz/cm²; the average degree of nucleon polarization (P); and, finally, the impact of the target magnet on particle detection, including restrictions on the detector solid angle and deflection of the particle trajectories by the target's magnetic field. The average nucleon polarization is

$$P = P_N \times D, \quad (2)$$

where P_N is the polarization degree of the polarizable nucleons, and D is a dilution factor, which takes into account the fraction of polarizable nucleons in the target material.

There are several experiments with polarized targets specifically designed to study the DIS/SIDIS processes:

1. Internal polarized-target experiment HERMES at the HERA storage ring at DESY operated at a low luminosity of $L \approx 1 \times 10^{32}$ Hz/cm². Both polarized-proton¹⁶ and polarized-neutron targets¹⁷ were developed. A polarization of protons $P_p \approx 0.9$ was achieved with a dilution factor $D = 1$. These targets allow an almost 4π solid angle for the detectors and require only a modest magnetic field of 3 kG on the target in spite of the high intensity of the electron beam.
2. High-energy, low-intensity muon-beam polarized-target experiments^{18,19}, which provide a modest luminosity of $L \approx 5 \times 10^{32}$ Hz/cm² (achieved with the beam intensity of 3×10^7 muons per second, target length of almost 1.5 m and total areal density of ~ 50 g/cm²), $P_p \approx 0.9$, and a dilution factor of $D \approx 0.16$. This lower dilution factor reflects a large fraction of unpolarized nucleons in that target. The magnetic field

on the target could be as low as 5 kG during data taking. Because of the low beam intensity, the target polarization lasts for hundreds of hours. Re-polarization of the target is performed in a 50 kG field, requires several hours, and is usually repeated every two days. A combination of the high beam energy and a modest value of the target's magnetic field allows for the required detector acceptance.

3. Polarized-target experiments with up to 32 GeV electron beams at the Stanford Linear Accelerator Center (SLAC)²⁰⁻²², which used a ^3He target²³ with $P_n \approx 0.3$, effective dilution $D \approx 0.125$, $L \approx 3 \times 10^{36}$ Hz/cm², and NH_3/LiD targets²⁴ with $P_p \approx 0.9$, $D \approx 0.16$, and $L \approx 7 \times 10^{35}$ Hz/cm² for studies of inclusive deep inelastic scattering.
4. There are several polarized targets for experiments with the high-intensity medium-energy electron beam at JLab²⁵:
 - i) An advanced version of the SLAC ^3He target with neutron polarization of $P_n \approx 0.43$ and $D \approx 0.25$. Beam intensities up to 15 μA were used resulting in $L \approx 3 \times 10^{36}$ Hz/cm². The latest development with circulating-gas approach allows even higher luminosity²⁶.
 - ii) A polarized NH_3/ND_3 target from the SLAC experiments and a similar one for the large-acceptance detector CLAS²⁷.
 - iii) A “frozen-spin” target for the CLAS detector²⁸.
 - iv) A novel HD polarized target for the CLAS detector. This target is designed to provide a high degree of nucleon polarization, a low dilution factor of 0.33, and a modest magnetic field on the target of 10 kG. The target²⁹ is projected to operate at a luminosity of $L \approx 1 \times 10^{34}$ Hz/cm².

The JLab accelerator will be upgraded to 12 GeV. A large set of polarized-target experiments (both DIS and SIDIS) is already approved, and some experiments are already in preparation³⁰. In addition to the upgrade of the polarized targets, there is a program developing a set of new detectors. It is apparent that, thanks to the advances in the ^3He target, investigation of the neutron structure has a good outlook. Higher-luminosity ^3He targets additionally allow operation in small magnetic fields (typically, 25 G).

C. Polarization and magnetic-field issues

Investigation of the nucleon spin structure requires the study of both inclusive and semi-inclusive processes, which means that average nucleon polarization is important, and that unpolarized nuclei in the target cause a loss of experimental accuracy. For example, in a polarized NH_3 target, the average nucleon polarization is only 1/6 of the free-proton polarization. All known targets with polarized protons and some with polarized neutrons operate at low temperatures, where the heat load from the incident particle beam becomes an issue, in addition to the radiation damage in the target material.

Magnetic fields required in proton polarized targets should be large, 50 kG, with a 10^{-4} uniformity. Such requirements result in limited acceptance usable for the experiments' detectors. A reduction of the field is possible for a low-luminosity regime. In many currently running and planned experiments, nuclear spins in the target should be oriented transversely to the beam direction. A transverse magnetic field bends the beam trajectory and leads to an additional background in the detector. Even more importantly, the field deflects the secondary particles of interest, whose energies are much lower than the incident-beam energy. This problem is significant for medium-energy experiments at JLab, where the secondary particles have energies in the 2-4 GeV range. With a transversely polarized NH_3 target, the deflection of a 2 GeV pion is about 15 degrees, which is too large for a SIDIS experiment.

D. Basic parameters of a target and luminosity

Productivity of an experiment depends on the target's Figure-of-Merit (FoM), which is defined by luminosity, target polarization, and the dilution factor as

$$FoM = L \times P^2 = L \times (P_N \times D)^2. \quad (3)$$

A minimum FoM required for a productive future SIDIS experiment, even with a large solid angle detector as CLAS, is about 10^{33} Hz/cm². With the two protons in a C_2H_4 molecule polarizable by the parahydrogen technique discussed below, a projected dilution factor is 0.07. As a result, the above FoM translates into a required luminosity of 2×10^{36} Hz/cm². If beam induced target warm-up and depolarization are not important, the beam intensity at JLab could be up to 100 μA . With such a large beam intensity, the required amount of material is small, 3 mg/cm². Assuming a target area of 1.5 cm², the total mass of the

material in the target is 5 mg. Using a conservative value for the spin-relaxation time of $T_1 \approx 10$ ms, one can see that the reactor should produce at least 500 mg/s of polarized C_2H_4 material. In this scheme, the power deposition in the target by the beam is ≈ 1 W, and the temperature rise is small due to a significant mass flow.

An ideal target for polarized-target experiments would (i) consist solely of protons (a simple nucleus) at high density and with (ii) full polarization. Moreover, it would allow for (iii) fast and efficient reversal of polarization (necessary to control systematic effects), (IV) would not require cryogenic temperatures to operate, and (V) would not require tesla-size magnetic fields hindering one's ability to control the polarization.

Here, we propose a concept of a polarized target that potentially addresses all of these requirements, with the exception of (i). The idea is based on the concept of parahydrogen induced polarization (PHIP) of nuclei^{31,32}, and in particular, its zero-field version³³.

II. THE CONCEPT OF THE TARGET

The basic idea of the proposed target is illustrated in Fig. 1. Pure parahydrogen (pH_2) is mixed with the initial substrate, for example, compressed ^{13}C -enriched acetylene (C_2H_2) gas or liquid, pre-loaded with an appropriate catalyst (Sec. IV,V). The mixing occurs in the Reactor, where a catalyst-assisted hydrogenation reaction takes place, adding the two hydrogen atoms across the carbon-carbon double bond.

Upon hydrogenation, the two protons coming from a parahydrogen molecule (and which are now incorporated in the product, for example, C_2H_4 molecule) are initially in the singlet state, however, the combined spin state of ^{13}C and the protons undergoes evolution under the scalar (J -coupling) Hamiltonian. This evolution results in a spin state that is devoid of net dipole polarization; however, such polarization can be induced³⁴ by applying an appropriate magnetic pulse (a typical pulse would be of DC magnetic induction ~ 1 G and a millisecond duration).

Once the hydrogenation reaction is complete, the product flows into the actual target volume. Once there, it is exposed to a DC pulse induced by the magnetic coil (shown in Fig. 1 surrounding the Polarized Target). After the DC pulse, proton polarization will oscillate with a characteristic J -coupling frequency on the order of 100 Hz, while undergoing relaxation with a typical decay time of several seconds. The direction of the polarization is

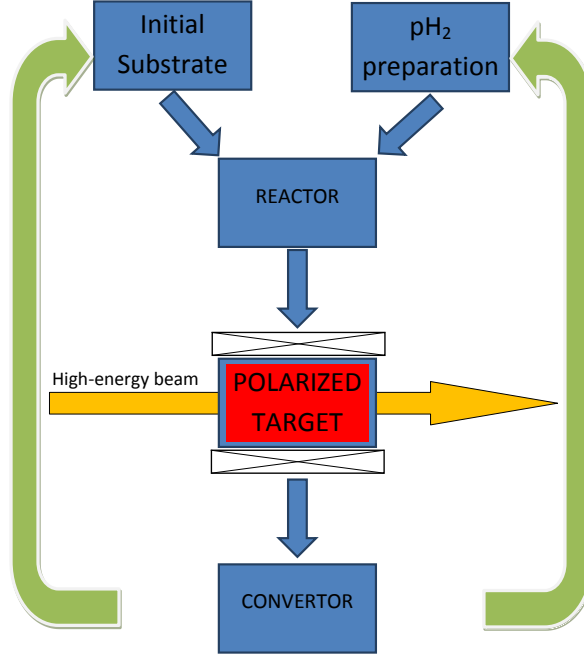


FIG. 1. A block diagram of the proposed arrangement.

collinear with the direction of the magnetic-field induction in the pulse. The polarization oscillation represents the fast reversal of the polarization in the target. Once the polarization oscillation decays, the contents of the target is emptied into the converter, while a fresh batch of hydrogenated material flows into the target volume. The used product is chemically regenerated (dehydrogenated) into substrate and hydrogen in the Converter, and can be reused for subsequent cycles.

III. *J*-COUPLING INDUCED POLARIZATION OSCILLATIONS

As mentioned above, a promising candidate for a target polarized via parahydrogen is ethylene (C_2H_4), synthesized from ^{13}C labeled acetylene ($^{13}\text{C}^{12}\text{CH}_2$) via addition of parahydrogen. Figure 2 shows the zero-field NMR spectrum of such a compound, following application of a DC pulse of magnetic field. After such a pulse, magnetization oscillates along the

direction of the pulse field at several frequencies, corresponding to rapid reversal of ^1H and ^{13}C spins with respect to each other. The different frequencies displayed here should also show up in a Fourier transform of the scattered signal. The zero-field NMR signal could thus be used as a diagnostic/reference in scattering experiments. We note that stray magnetic fields, including the magnetic field of the high-energy beam itself may modify the spectrum, so it will be important to control the fields at a milligauss level.

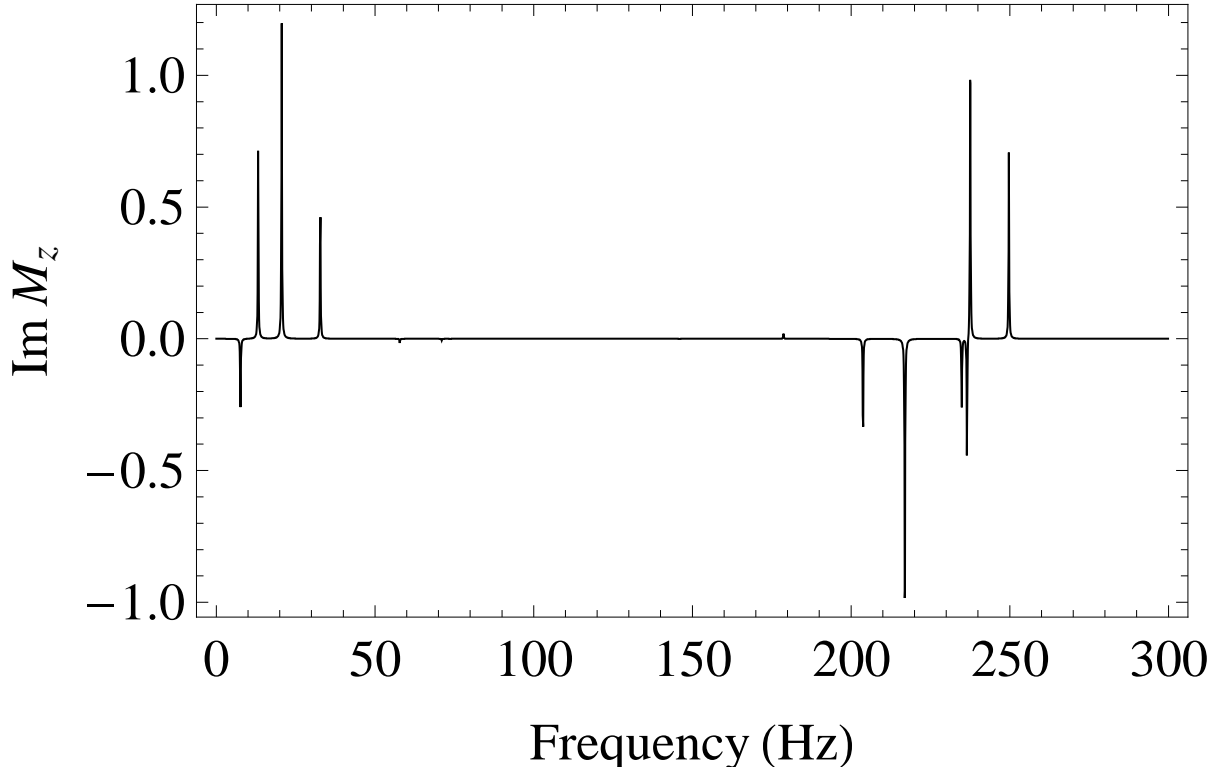


FIG. 2. Imaginary component of the zero-field Phip enhanced spectrum of ethylene, synthesized from singly ^{13}C labeled acetylene via addition of parahydrogen. This simulation shows oscillations of the observable nuclear magnetization at several frequencies corresponding to relative reversal of proton and ^{13}C spins.

IV. HYDROGENATION AND DEHYDROGENATION REACTIONS

Parahydrogen-induced polarization (PHIP) proceeds by hydrogenation across an unsaturated carbon-carbon bond. For example, propene (C_3H_6) can be hydrogenated to yield propane (C_3H_8) using a suitable catalyst. Recently, it has been demonstrated for this re-

action in the gas phase³⁵ that it is possible to obtain up to 60% polarization at the sites of the two added protons. This means that the overall polarization of the hydrogen nuclei on the substrate molecules is $\approx 0.6 \times (2/8) = 15\%$. The theoretical maximum (for 100% PHIP) would be 25% overall polarization. An improved choice may be ethylene (C_2H_4) to ethane (C_2H_6), which would yield a theoretical maximum of $\approx 33\%$. If one uses acetylene (C_2H_2) as the starting molecule, such a reaction will yield a mixture of ethylene and ethane. The interesting product here would be polarized ethylene, with four hydrogen nuclei: two of which are polarized and two are unpolarized, yielding a potential theoretical maximum polarization of 50%. It may be possible to separate the desired product from the mixture and condense it to the liquid phase using a liquid nitrogen [boiling point (b.p.) 77 K] cold finger to get a high density of polarized hydrogen nuclei (b.p. of ethylene is 169.5 K). We note that one way to obtain nuclear-spin order that can be converted into magnetic polarization in zero or low magnetic fields is to use a molecule that possesses an asymmetric J -coupling topology. In the case of propane, it is sufficient that one of the carbons adjacent to the added protons be magnetic (e.g., ^{13}C). Subsequent evolution of the spins of the protons and the magnetic nucleus under the influence of each other leads to net magnetization that can be generated after the application of a magnetic-field pulse.

Because selectively ^{13}C -labeled molecules are expensive and experiments done in continuous-flow mode may rapidly consume molecules, it may be desirable to recycle the hydrogenated product (see Fig. 1). To this end, we may envisage closing the circuit by adding a dehydrogenation step after the polarized target loses its polarization. Dehydrogenation of propane and other alkanes is possible, for example, with the use of vanadium, molybdenum and tungsten oxides³⁶, albeit catalysts will differ in their selectivity and the conditions of the dehydrogenation reaction (pressure, temperature) may be different.

V. OPTIMIZATION

In order to make an efficient polarized target, all aspects of the process must be optimized. The following is a partial list of parameters and issues that should be addressed.

- The choice of the substrate molecule. Our goal here is to have the highest fraction of polarized nucleons compared to the overall number of nuclides in the molecule in addition to having an overall high areal density of the target. The efficiency of

polarization, polarization relaxation time (which we need as long as possible), the ability to flow and recycle the material are all important factors. Will the substrate be compressed gas or liquid, or, perhaps, a supercritical phase? Liquid state would be advantageous in terms of density and, typically, longer relaxation times than gas; however, elevated pressures and/or low operating temperatures may be required to suppress evaporation. An additional consideration is the temperature dependence of the hydrogenation reaction.

- The choice of the catalyst that will ensure efficient hydrogenation on the time scale of spin relaxation. This is essential to have high spin polarization. Both homogenous and heterogeneous catalysts options are available. Heterogeneous catalysts have the advantage that they can be packed into a reactor. The reacted product is free of catalyst contamination, which would be detrimental for this experiment. There is also no need for a solvent in many heterogeneous reactions. The highly efficient catalyst for PHIP³⁵ mentioned above meets these requirements.
- The converter chemistry: If a heterogeneous catalyst is used, a catalytic converter (reactor) is needed. There are important issues that must be addressed when designing an appropriate reactor. The reactor should be dense enough for efficient reaction, but not too dense as to hinder transport processes. Efficient heat transport is required to avoid deterioration of the catalyst. A short residence time is desirable in order to avoid depolarization of the product.
- An efficient flow/shuttle/transport system is required to bring the reaction product to the target region. Short transit times are needed to avoid depolarization. Sufficiently good control of the transport is needed to transport and subsequently immobilize the product.

VI. CONCLUSION AND POSSIBLE EXTENSIONS

A spin-polarized-target concept based on parahydrogen-induced polarization (PHIP) has been proposed. The PHIP targets may offer a number of advantages over the existing polarized targets, including fast polarization reversal and non-cryogenic operation at near-zero magnetic field. So far, no “show stoppers” are seen; however, specific technical details

of the novel target still need to be worked out, which is partially a chemical-engineering task to be addressed in future work.

There are several interesting extensions of the basic concept that deserve further consideration:

- Nonhydrogenative (NH) PHIP^{37,38} expands the types of substrates that could be polarized, and, more importantly, eliminates the need for the dehydrogenation step. Note that a zero-field nonhydrogenative PHIP experiment has recently been performed³⁹. It is interesting to note in this context that in the work⁴⁰, where NH-PHIP of aminoacids and peptides was demonstrated, hyperpolarized HD molecules were also produced, which are an attractive polarized-target material.
- The PASADENA/ALTADENA PHIP schemes (see, for example, the review³²) that rely on relatively high magnetic field and removing the degeneracy between the two protons not by J -coupling, but by chemical-shift differences, may also be considered. The advantage here could be that protons are the only polarized nuclei (for example, no ^{13}C nuclei are involved).
- The possibility of doing the hydrogenation reaction in-situ should also be considered, which will likely involve homogeneous catalysis. An advantage is that this would avoid sample transfer.
- Photochemically induced reactions with parahydrogen should be considered. More generally, photochemically induced nuclear polarization (CIDNP⁴¹, without parahydrogen) that has been discussed in the context of polarized targets for over twenty years⁴² should be revisited.
- Finally, we briefly discuss PHIP polarized ethyl fluoride $^{12}\text{CH}_3$ - $^{12}\text{CH}_2$ - ^{19}F . The initial pH_2 singlet polarization at zero field “spreads” over the whole molecule via the J -coupling network. The difference with the cases discussed in Sec. IV, is that here, instead of ^{13}C , it is ^{19}F that breaks the symmetry between the protons, as revealed by application of the magnetic-field pulse. In this case, however, the oscillation frequency after the pulse is smaller due to the fact that the second-order H-F coupling is smaller than the first-order H- ^{13}C coupling ($^2J_{\text{HF}} < ^1J_{\text{HC}}$). Now, the ^1H chemical shift of $^{12}\text{CH}_3$ is different from that of $^{12}\text{CH}_2$ -F (by a few ppm), so at moderate DC

magnetic fields ~ 1000 G, after the pulse, the magnetization will oscillate with the Larmor frequency of ^1H and ^{19}F (modulated by the J -coupling). This means a much faster reversal of the polarization (up to several MHz), but at a cost of applying a homogeneous magnetic field of ~ 1000 G.

Finally, we note that the development of efficient nuclear polarization techniques may have important ramifications also outside of the polarized-target context, including the entire plethora of the applications of NMR and magnetic-resonance imaging, production of polarized proton beams, the studies of β -decay asymmetries, and in enhancement of nuclear-fusion reactions via nuclear polarization⁴³.

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